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Thermal and laser conditioning of production- and rapid-growth KDP and KD*P crystals

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ABSTRACT

Large solid state lasers such as Beamlet and the proposed National Ignition Facility (NIF) require optical materials with extremely high damage thresholds. Potassium dihydrogen phosphate (KDP) and its deuterated analog (KD*P) both require some form of conditioning to reach the design fluence of these lasers. Both the bulk material and the crystal surfaces must have damage thresholds in excess of 16 J/cm^2 at 1053 nm and 11 J/cm^2 at 351 nm for 3-ns pulselengths. The use of ultrafiltration techniques has been demonstrated to produce bulk material with damage thresholds exceeding these requirements with the use of R:1 laser conditioning. More recent results at LLNL using large-area laser conditioning and thermal annealing are described for a variety of state-of-the-art KDP and KD*P crystals. Results on thermally annealed KD*P with a deuteration range of 60% to 80% are also presented, and compared to those of ordinary KDP.

1. INTRODUCTION

Large, high-damage-threshold KDP (KH_2PO_4) and KD*P ($KD_{2x}H_{2(1-x)}PO_4$, $x \geq 0.6$) crystals are required for high-peak-power lasers in the Inertial Confinement Fusion (ICF) Program at Lawrence Livermore National Laboratory (LLNL). KDP crystals 27 x 27 cm^2 in size have been used since 1984 on the Nova laser at LLNL for harmonic conversion from the 1053-nm fundamental to the 351-nm third harmonic using the Type II/Type II scheme. LLNL is currently building the Beamlet laser, a scientific prototype of the proposed 240 beam National Ignition Facility (NIF). Beamlet and the NIF both require larger (37-40 cm) KDP and KD*P crystals than are currently employed in Nova. The damage threshold of these crystals must also be approximately two times higher than required for Nova, on an equivalent pulse length basis.

Because of their multipass architecture, Beamlet and the NIF require a large KDP or KD*P crystal in a full-aperture electro-optic switch.[1] This longitudinal Pockels cell uses a moderate-density helium plasma (transparent at 1 µm) to form electrodes on the crystal faces. The plasma-electrode Pockels cell to be used in the Beamlet laser will use a 37-cm crystal. NIF is currently projected to require a 40-cm KD*P or KDP crystal. The crystal composition will be determined by trading off the lower cost and higher 1-µm absorption of KDP (which requires a slightly larger injection energy from the pulse generation laser) against an increased cost and reduced absorption of KD*P. Performance on Beamlet will be validated with both KDP and KD*P crystals.

The baseline frequency conversion scheme is Type I/Type II on Beamlet and the NIF, utilizing a Type-I KDP crystal followed by a Type-II KD*P crystal. KD*P, although more expensive that KDP, has a lower spontaneous Raman cross-section than KDP. At the higher intensities and larger apertures of Beamlet and NIF, relative to Nova, stimulated Raman scattering (SRS) is expected to be a significant loss at the output of the tripler.[2] Hence, only the third harmonic crystal is deuterated. Experiments on Beamlet in 1994-95 will be used to verify the theoretical analysis and confirm the requirement of KD*P. The use of KD*P has significant implications with respect to the damage threshold at 3ω, discussed in Section 3.3.

The estimated average and peak 3-ns fluences for the final pass of the Beamlet laser are plotted in Figure 1; NIF fluences are comparable. The crystal requirements are summarized in Table 1.

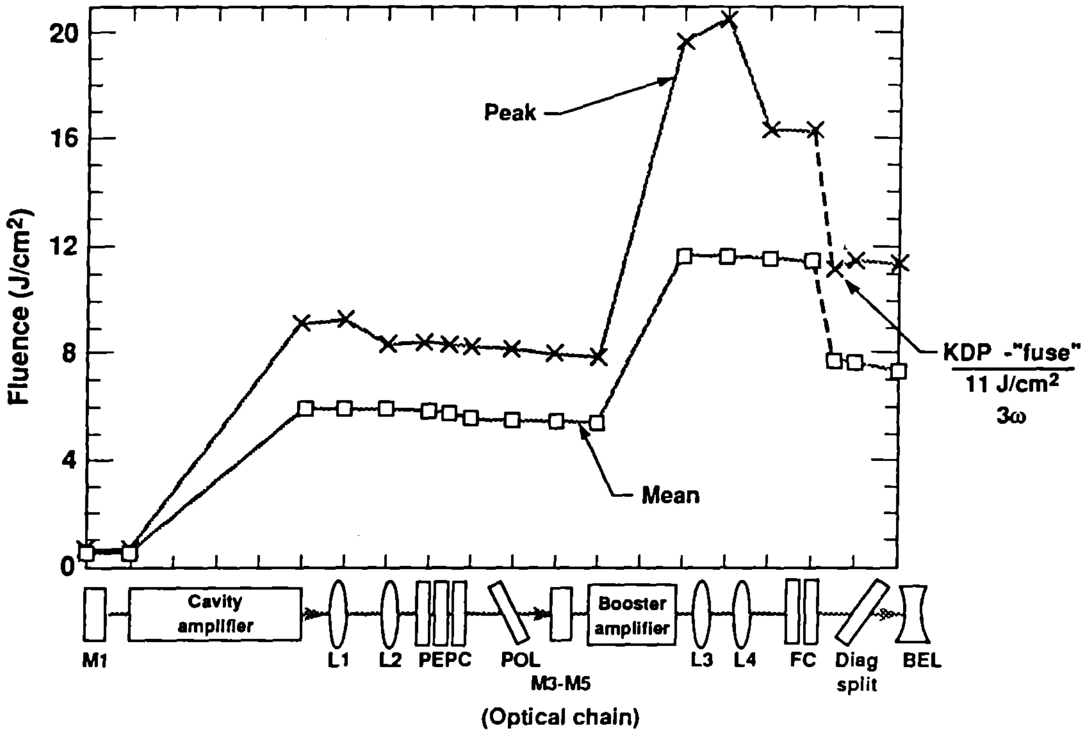

Figure 1. Estimated average and peak fluences for the final pass on the Beamlet laser. The peak fluences in the crystals are 16 J/cm^2 at 1ω and 11 J/cm^2 at 3ω.

Table 1: Estimated fluences and required damage thresholds in the Pockels cell, second harmonic generation (SHG), and third harmonic generation (THG) crystals (3-ns pulse)

		Fluences (J/cm^2)		
Location	Frequency	Average	Peak	Desired Threshold
Pockels cell	1ω	6	8.5	10
SHG	1w	11.5	16	19
THG	3ω	8	11	13

Rainer et al.[3] showed that the measured unconditioned damage threshold in KDP and KD*P nominally exhibits an $\omega^{1/2}$ dependence on laser frequency. When scaled from 3ω to 1ω, the desired damage threshold for the THG crystal becomes 22 J/cm^2. Thus, the damage threshold of the KD*P crystal at 3ω is expected to be the limiting factor (the *fuse*) with respect to damage. (A higher fluence is not useful because the peak conversion efficiency occurs at about 3-4 GW/cm2 (1ω), which corresponds to 9 to 12 J/cm^2 average fluence for a 3-ns pulse.[4] For this reason, the Beamlet 1ω average fluence has been set at 11.5 J/cm^2).

The peak fluences expected in these crystals generally exceed the unconditioned damage threshold,[3] particularly at 351 nm. Therefore, some form of conditioning is required to raise the damage threshold above the peak intensities in these positions. In Section 2, we briefly describe the laser parameters and conditioning approaches used to increase the damage threshold of KDP and KD*P crystals. In Section 3, we present and discuss the results for laser conditioning (3.1) and thermal conditioning of KDP (3.2) and KD*P (3.3).

2. EXPERIMENTAL PARAMETERS

Most of the damage-threshold measurements were made with the Chameleon laser facility at LLNL. We conducted our tests with nominal 3-ns pulses (full-width, half-maximum Gaussian shapes) at 355 and 1064 nm. Samples were irradiated at a pulse-repetition frequency of 10 Hz with 600 shots unless massive damage was observed earlier. The beams had smooth Gaussian profiles with 1/e2 diameters > 1 mm. The experimental configuration for testing KDP crystals is described in detail in Reference 3.

We conducted damage tests and performed conditioning of the crystals by a variety of techniques. These are described in Figure 2. The primary test methods are with S:1 irradiation (600 shots at the same fluence) and R:1 irradiation (600 shots ramped up in fluence). These methods formed the basis for our respective unconditioned and conditioned damage-threshold measurements. Rastering a sample through a fixed laser beam to condition it was accomplished on a small scale (≤ 50-mm scans) using the Reptile, Raster Blaster, or Thor laser facilities.[5] This can be accomplished on full-sized crystals (~ 40-cm scans) with the Plato facility.[6] The final conditioning mechanism, thermal annealing, was done off-line in a controlled oven environment.

3. CRYSTAL CONDITIONING

3.1 Laser conditioning of KDP and KD*P

The ability of laser conditioning to increase damage threshold has been established for over a decade. During this time, both the conditioned and unconditioned damage thresholds have increased significantly (by about a factor of 2).[5] Even with this sizeable improvement in damage threshold, laser conditioning still raises the damage threshold at both 1ω and 3ω by an average factor of 2.1 for samples measured in the last 2 years (Figure 3). Although the damage mecha-

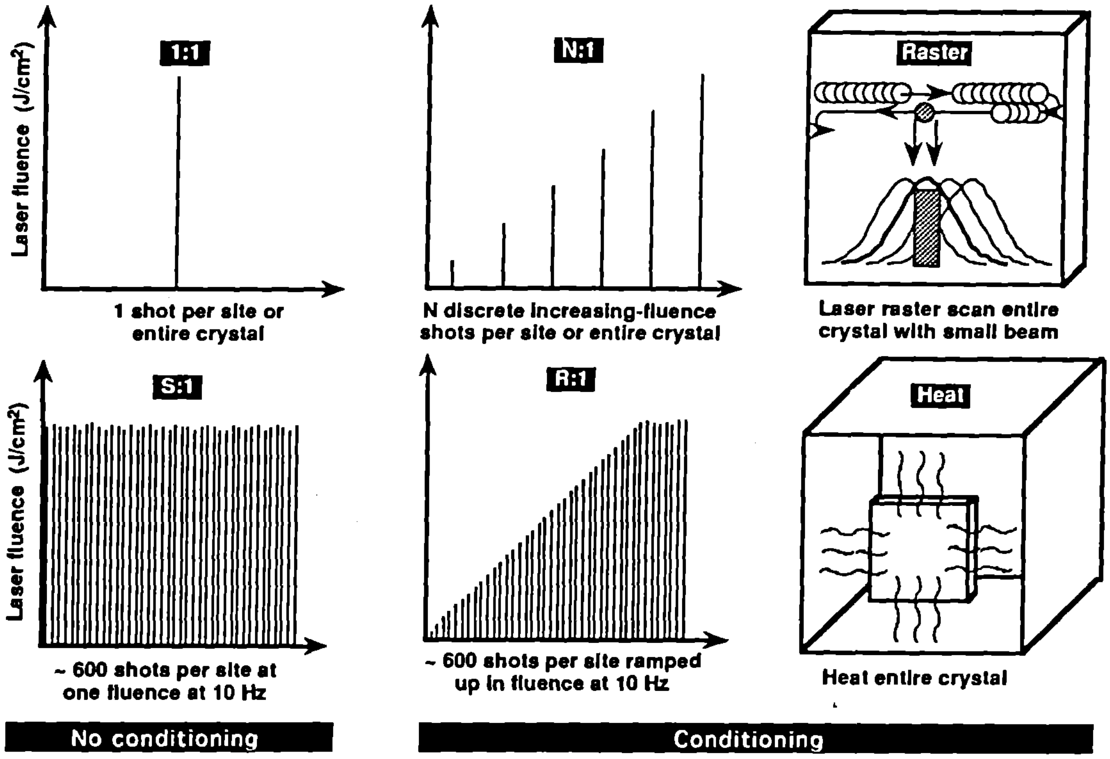

Figure 2. Types of laser irradiation and conditioning methods.

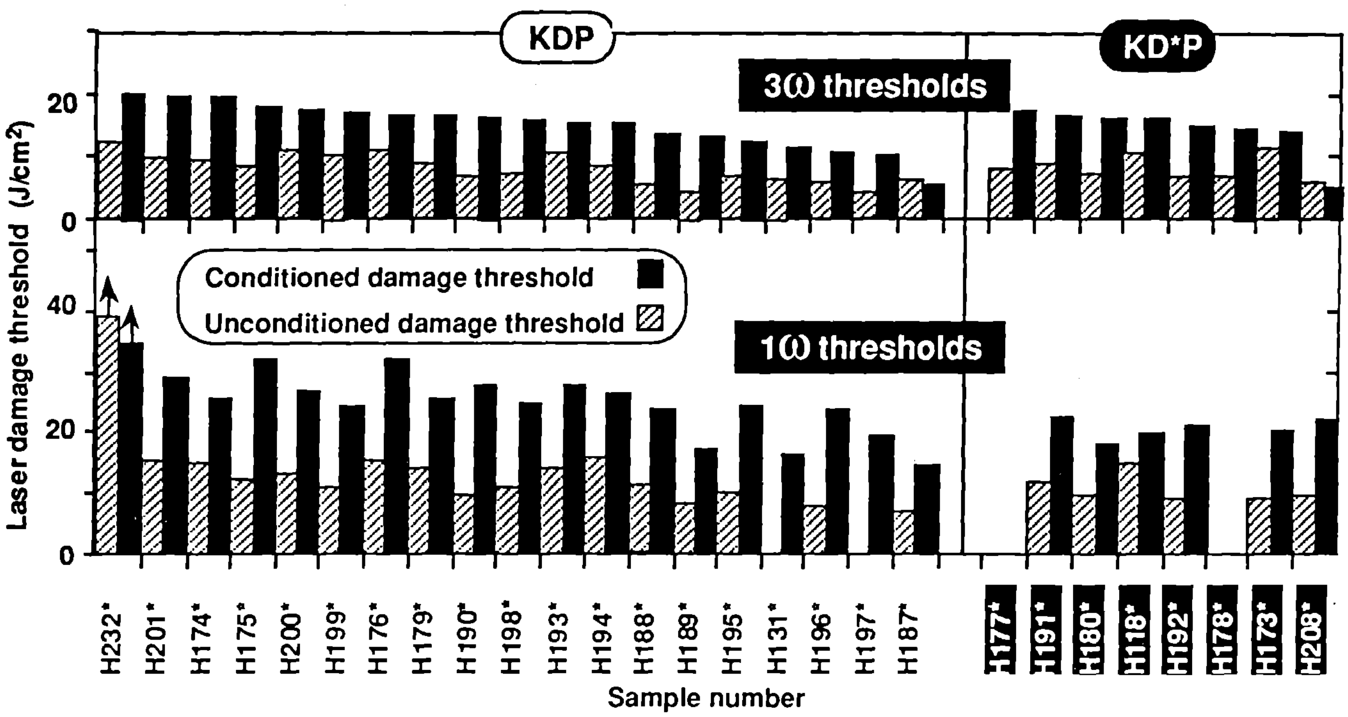

Figure 3. Effect of laser conditioning (R:1) on damage threshold. Laser conditioning raises the 3-ns thresholds by an average factor of 2.1 at 1ω and 3ω.

nism for bulk KDP hasn't been established conclusively, the pulse length scaling of $\tau^{1/2}$ is consistent with thermal absorption and dissipation.[3] Only the lowest damage-threshold samples of KDP and KD*P have failed to show substantial improvement in 3ω damage threshold by laser conditioning (see H187* and H208* in Figure 3). Even these poorer samples show significant damage threshold increases at 1ω. Of comparable significance to the improvement in damage threshold with laser conditioning is a reduction in damage severity. The damage sites generally are smaller in laser conditioned crystals (~ 10-30 μm versus 50-200 μm) and do not grow with repeated shots at the threshold fluence.

Although laser conditioning has been shown to improve the damage threshold, small-spot, R:1 conditioning (several hundred shots in a ramped sequence) is impractical for the large crystals necessary for Beamlet and NIF. Conditioning of thin-film HRs and polarizers has been demonstrated using 6-10 steps (N:1) in the last few years.[7] This approach is being used to condition HRs and polarizers for Beamlet, by rastering a small beam over the entire aperture at successively higher fluences as illustrated in Figure 2.[6] We have initiated a study to determine the feasibility of raster conditioning bulk KDP crystals. The results of Figure 4 show that raster conditioning KDP improves bulk damage thresholds at 1ω and 3ω above S:1 levels, but below R:1 values. The improvement at 1ω is substantially above the Beamlet requirement of 16 J/cm^2. At 3ω, the improvements were mixed. Sample H194* showed noticeable 3ω improvement with conditioning at 1ω.

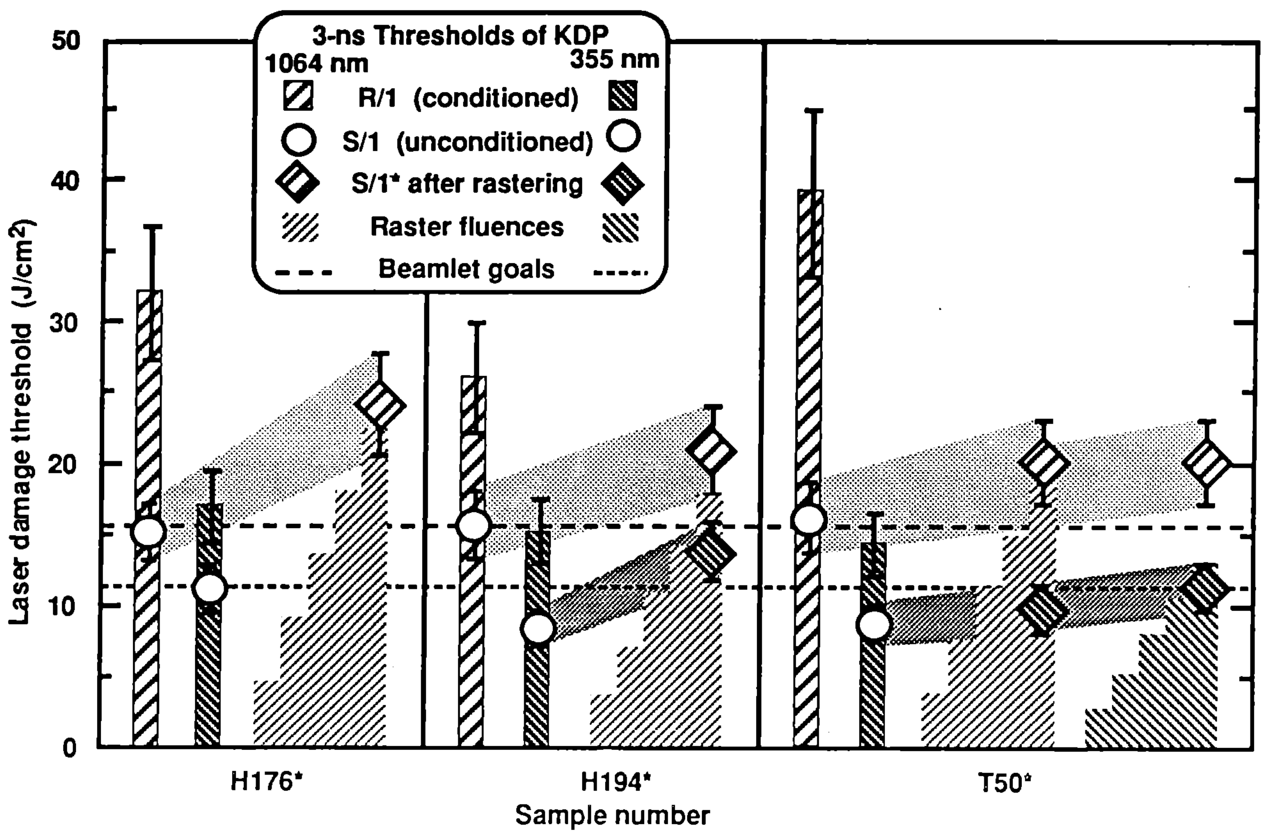

Figure 4. Effect of raster irradiation on damage threshold. Raster irradiation of a large area with 5 shots per site using a small beam provides partial conditioning of KDP.

Sample T50* was conditioned at 1ω, and the damage threshold measured at both 1ω and 3ω. The improvement at 3ω over the S:1 threshold was marginal, and below the Beamlet goal. Conditioning with 3ω did raise the 3ω threshold slightly. These preliminary results suggest that raster conditioning of KDP crystals in 5 steps with a 1 micron beam is acceptable for 1ω damage, but probably not for 3ω. Thermal conditioning (discussed in 3.2 and 3.3) shows greater promise for improving 3ω damage threshold, especially for KDP. Should thermal conditioning of KD*P prove ineffective at 3ω, the present plan is to condition the Beamlet KD*P THG crystals on-line at full aperture using approximately 10 steps. This issue is discussed further in Section 3.3.

3.2 Thermal conditioning of KDP

Thermal conditioning was investigated for improving KDP damage threshold in the late 1970s and early 1980s at LLNL and elsewhere.[8,9] At LLNL, thermal conditioning was investigated at 140°C, and shown to improve the 1ω damage threshold somewhat above the 1:1 (1 shot per site) value, but below the N:1 level.

The temperature range over which KDP conditioning can be investigated is limited by the destructive tetragonal/monoclinic phase transition which occurs at about 180°C. We performed our initial experiments at 165°C - 175°C, just below the phase transition temperature, and maintained this temperature for 48 hours. As can be seen in Figure 5, thermal conditioning of KDP at

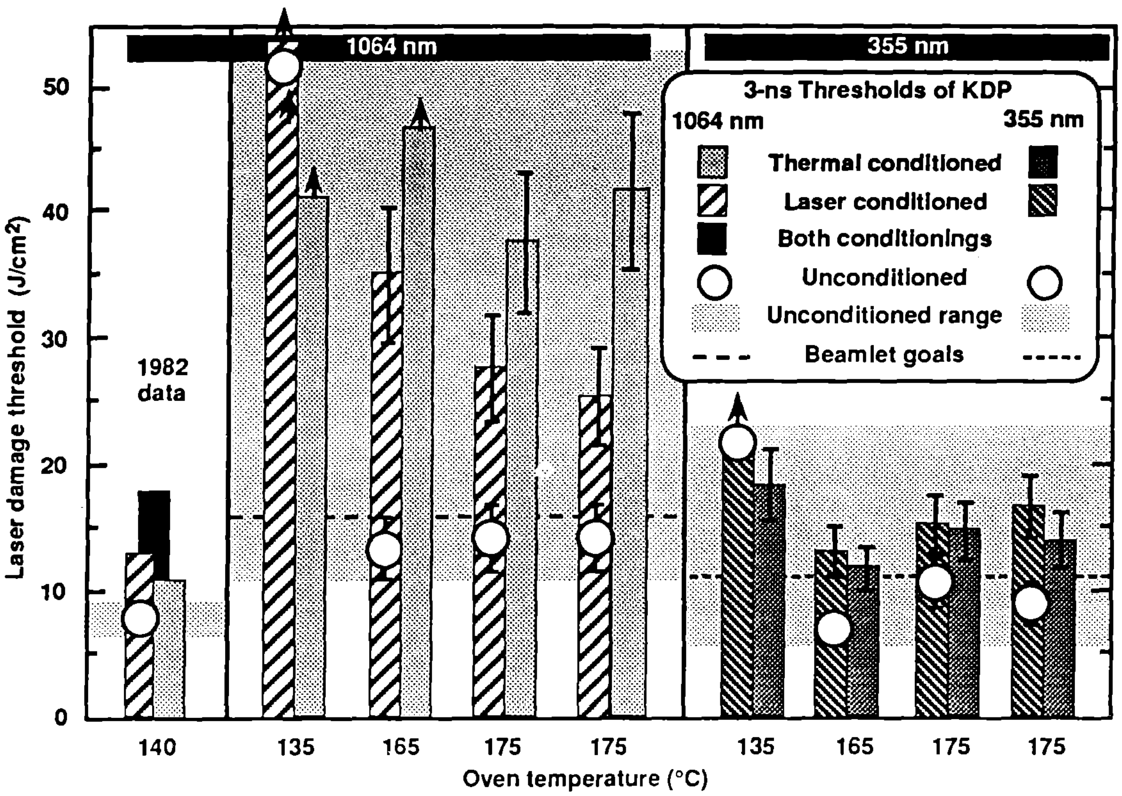

Figure 5. Comparison of laser and thermal conditioning of KDP. Thermal conditioning raises the bulk damage threshold of KDP above the laser conditioned value at 1ω, and is essentially comparable to laser conditioning at 3ω.

these conditions dramatically improved the 1ω damage threshold. The S:1 threshold after annealing was even higher than the laser conditioned threshold for three of the four samples measured.

The conditioning improvement at 3ω was not nearly as pronounced; in fact, the thermal conditioning was somewhat less effective than laser conditioning (but within error bars). We subsequently tested another KDP crystal conditioned at 135°C. This temperature was chosen because the silicone coating used in conjunction with the sol gel AR is cured at approximately this level. However, for this experiment, the pre-conditioned S:1 damage threshold was so high that it exceeded the peak laser-fluence capability. In fact, the unconditioned threshold of this particular crystal exceeded both the 1ω and 3ω damage requirements of Beamlet and the NIF. We hope to understand the connection between damage in this crystal and the conditioned crystals. We also plan to examine other KDP crystals in the range of 140°C, and for varying annealing times to investigate the kinetics of this process.

3.3 Thermal conditioning of KD*P

Based on successful results in thermally conditioning KDP to improve its damage threshold, we investigated the feasibility of increasing the damage threshold of 60-80% deuterated KD*P. The Beamlet THG crystal is 80% KD*P; 60% KD*P is being evaluated for the NIF (to reduce the crystal cost without impacting system performance). The tetragonal/monoclinic phase-transition temperature for KD*P is lower than KDP, varying inversely with the deuteration level. Highly deuterated KD*P (99%) undergoes a phase transition at about 100°C.[10] We found in this work that the phase transition temperature of 80% KD*P is about 130°C. This lower critical temperature may impact both the thermodynamics and kinetics of the conditioning process, assuming that the mechanism for KD*P is similar to KDP. The conditioning rate is expected to be slower at lower temperatures, assuming that an Arrhenius-type kinetic constant governs the conditioning process. The lower phase transition temperature of KD*P may also prohibit reaching a second critical temperature necessary to alter the defect(s) responsible for the observed laser damage.

The conditioning time for KD*P was fixed at 48 hours for comparison with the KDP results. The annealing temperature was varied between 110°C and 160°C for the 80% KD*P, and between 125°C and 135°C for the 60% KD*P. All of the 80% KD*P and 60% KD*P crystals were from the same two boules, respectively. For these conditions, Figure 6 shows that the 1ω damage threshold was improved by varying degrees over the unconditioned S:1 level, although the relative improvement at 1ω for KD*P was lower than for KDP. The 3ω damage thresholds were not significantly different from the S:1 unconditioned values. The 60% KD*P showed similar results to the 80%-KD*P material. We have not yet determined the phase transition temperature of 60% KD*P, but expect it to be in the range of 145°C to 150°C. We plan to examine the effect of annealing time on damage threshold in the near future to determine if longer exposure at temperature will improve the damage threshold, particularly at 3ω.

One interesting result was obtained for an 80%-KD*P crystal that was heated above the phase transition temperature to a metastable state at 160°C. This crystal exhibited a significantly improved damage threshold at 1ω, and a moderately improved level at 3ω. This increase in dam-

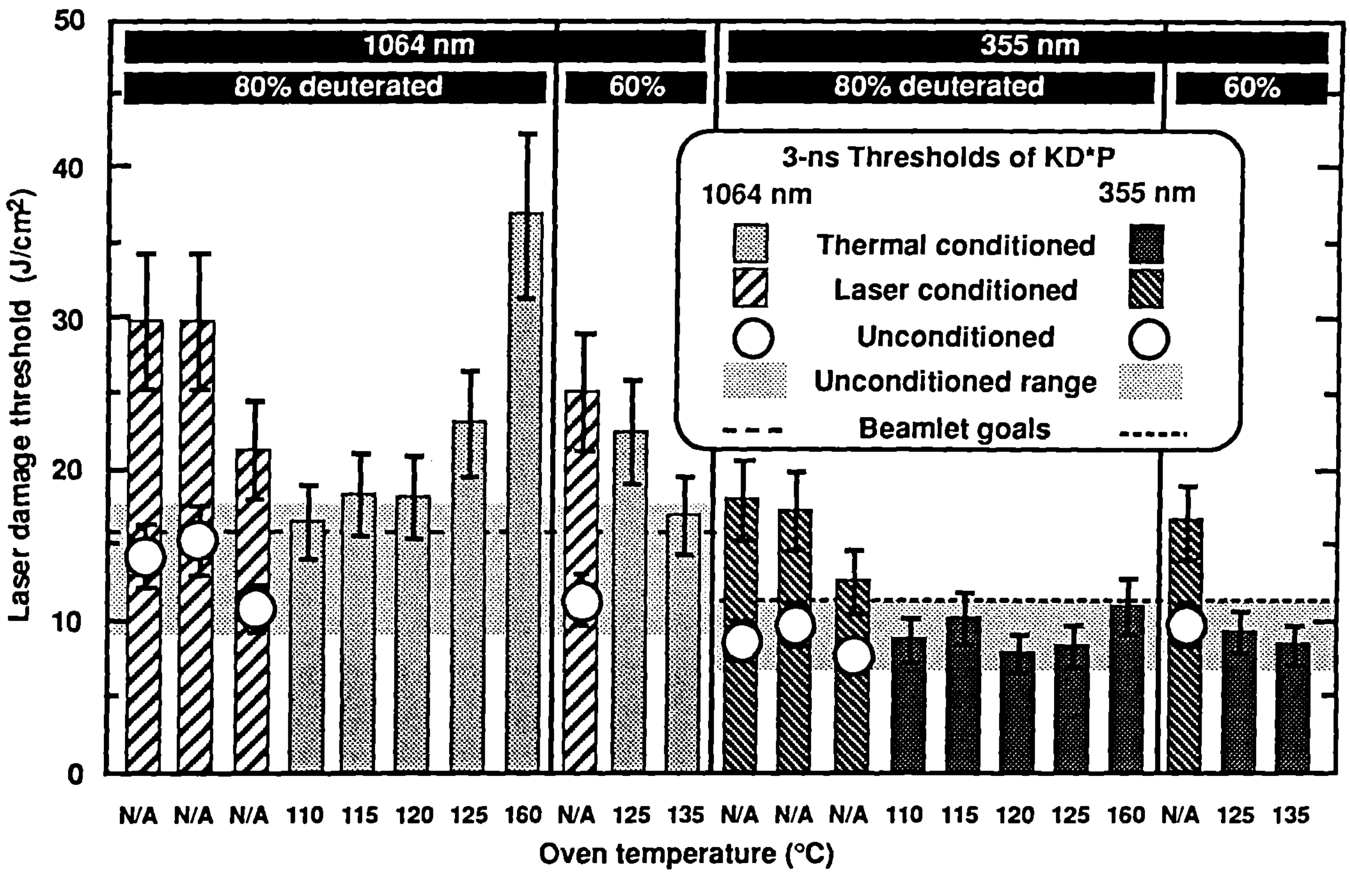

Figure 6. Comparison of laser and thermal conditioning of KD*P. Thermal conditioning improves the bulk damage threshold at 1ω to a level comparable to laser conditiong. At 3ω, no conditioning effect was observed for crystals heat treated below the monoclinic phase transition temperature.

age threshold is consistent with either a kinetic or thermodynamic limitation to damage threshold improvement. The experiments with longer annealing times will further elucidate the conditioning mechanism.

Figure 6 shows thresholds based primarily on the onset of subtle damage consisting of infrequent pinpoints. Figure 7 shows the same test data but with thresholds defined by major, but still acceptable, morphology for large laser systems (≤ 5 pinpoints, ≤ 50 μm in size). For this relaxed definition of damage threshold, thermal conditioning may marginally meet the Beamlet requirements at 3ω. Figure 7 also shows that laser conditioning after thermal conditioning further improved the damage threshold at 3ω. While not at all conclusive, it is consistent with the possibility that longer conditioning times could produce higher 3ω damage thresholds. Based on this result, our plan for Beamlet is to thermally condition the KD*P crystals for a yet to be determined period, which as a minimum should improve the damage morphology, and may improve the threshold above the 3ω requirement. On-line laser conditioning over about 10 increments will be used to further condition the crystals.

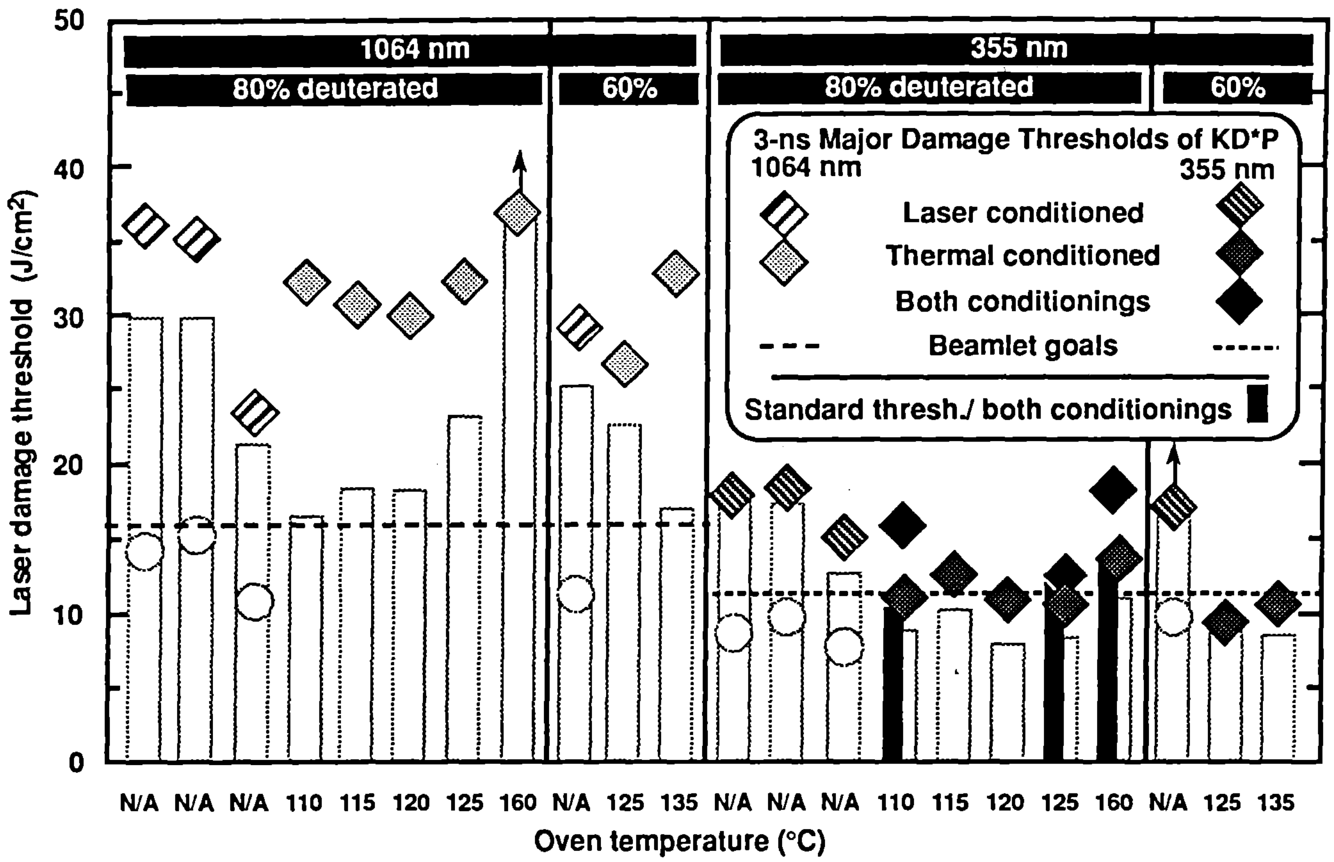

Figure 7. Bulk damage thresholds in KD*P crystals for a relaxed definition of damage threshold (≤ 5 pinpoints, ≤ 50 μm in size). The Beamlet goal for damage threshold for subtle but acceptable damage morphologies can still be acheived by thermal conditioning of KD*P.

4. ACKNOWLEDGMENTS

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5. REFERENCES

1. J.R. Murray, et al., "Upgrade of the LLNL Nova laser for inertial confinement fusion," *Solid State Lasers II*, SPIE Vol. 1410, pp. 28-39, January 1991.

2. R.A. Sacks, et al., "Stimulated Raman scattering in large-aperture, high-fluence frequency-conversion crystals," *ICF Quarterly Report*, UCRL-LR-105821-92-4, pp. 179-188, September 1992.

3. F. Rainer, et al., "Laser damage to production- and research-grade KDP crystals," *Laser-Induced Damage in Optical Materials: 1992*, SPIE Vol. 1848, pp. 46-58, October 1992.

4. C.E. Barker, et al., "High fluence third harmonic generation," *ICF Quarterly Report*, UCRL-LR-105821-93-2, pp. 55-62, March 1993.

5. F. Rainer, et al., "A historical perspective on fifteen years of laser damage thresholds at LLNL," *Laser-Induced Damage in Optical Materials: 1993*, to be published.

6. L. Sheehan, et al., "Large-area conditioning of optics for high-power laser system," *Laser-Induced Damage in Optical Materials: 1993*, to be published.

7. M.R. Kozlowski, et al., "Laser conditioning and electronic defects of HfO2 and SiO2 thin films," *Laser-Induced Damage in Optical Materials: 1990*, SPIE Vol. 1441, pp. 269-282, October 1990.

8. J. Swain, et al., "The effect of baking and pulsed laser irradiation on the damage threshold of potassium dihydrogen phosphate crystals," *Appl. Phys. Lett.* Vol. 41, pp. 12, 1982.

9. V.V. Azarov et al., "Effect of annealing on optical homogeneity of potassium dihydrogen phosphate single crystals," *Izv. An. SSSR*, Vol. 18, pp. 164-165, 1983.

10. L.N. Rashkovich, KDP-family single crystals, pp. 14-19, Adam Hilger, New York, 1991.